

Removal of H₂S from Syn-gas Streams via an Electrochemical Membrane System
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An electrochemical cell to be used for desulfurization of coal gasification process streams is being tested at lab scale. Syn-gas is passed by the cathode of the cell, run at typical gasifier temperatures, where the H₂S is preferentially reduced to hydrogen gas, which enriches the fuel gas, and sulfide ions, which enter the electrolyte-filled membrane. These sulfide ions are oxidized to elemental sulfur vapor at the anode electrode, collected downstream, providing continuous performance. A gas mixture reflective of coal gasification process streams is fed into the electrochemical cell, which is kept at temperature by a custom-made furnace. At 600° C, after hydration and shift reaction, the fuel gas composition entering the cell is 34% CO, 22% CO₂, 35% H₂, 8.5% H₂O, and 450 ppm H₂S. Gas chromatography is used to monitor H₂S concentrations into and out of the cell. Infrared spectroscopy is used to monitor CO₂.

The key to optimum performance is the cathode material, which is exposed to the sour gas. Previous work under UCR sponsorship has shown that nickel and cobalt electrodes are operational but have some limitations in H₂S concentration and temperature. We have recently tested a new ceramic material that appears stable and conductive over large ranges in concentration as well as temperature. Full-cell work has begun utilizing this material, as well as comparison with known materials.

Results will be presented for the performance of three different cathode materials: Co₉S₈, LiY_(0.9)Ca_(0.1)FeO₃, and NiS. Operation temperatures were varied from 580 to 650° C while system pressure remained constant at 1 atm. The other electrochemical cell components consist of a lithiated nickel anode, an inert ceramic membrane (yttria-stabilized zirconia), steel housing (passivated by an aluminum layer), and (Li_{0.62}K_{0.38})CO₃ molten electrolyte held within the membrane. Quantitatively, over 90% removal of H₂S has been demonstrated, although long-term transport performance at these levels is yet to be proven. Electrical polarization is also excellent, with far less than one volt needed at all currents.

The removal rate of H₂S is limited by bulk mass transfer from the coal-gas stream to the cathode and migration of sulfur ions through the electrolytic membrane. Operating temperature and cell design parameters (i.e. membrane thickness, membrane tortuosity, and flow channel design) determine which of these processes limits the maximum current allowable for H₂S removal. Higher temperatures favor higher migration rates while thicker, more tortuous membranes decrease migration rate.

Lost current efficiency due to side reactions are of little concern because the power density is small in this process; however, loss of electrolyte could be a concern if carbonate is extensively reacted to form CO₂ and O₂ at the anode. In this regard, testing is being carried out with clean coal-gas. These tests will assure that stoichiometric transport of CO₂ can be achieved in either direction; that is, in the standard fuel-cell mode, from cathode to anode, or in the concentration-cell mode, from anode to cathode.

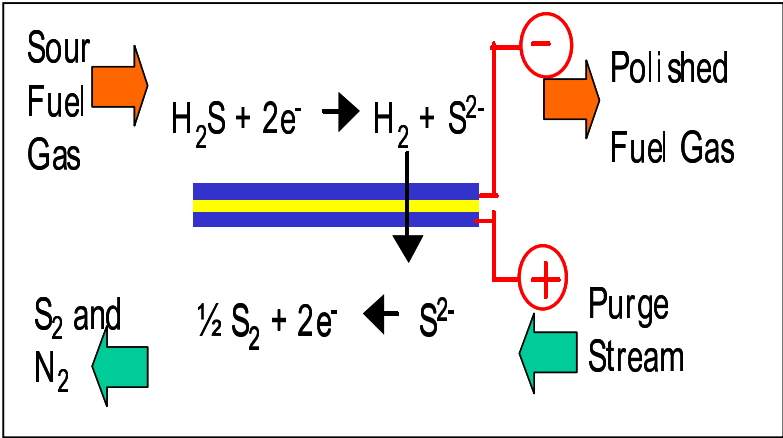


Figure 1. Schematic of sulfur ion adsorption at the cathode and migration across electrolytic membrane to the anode, where it is oxidized to elemental sulfur

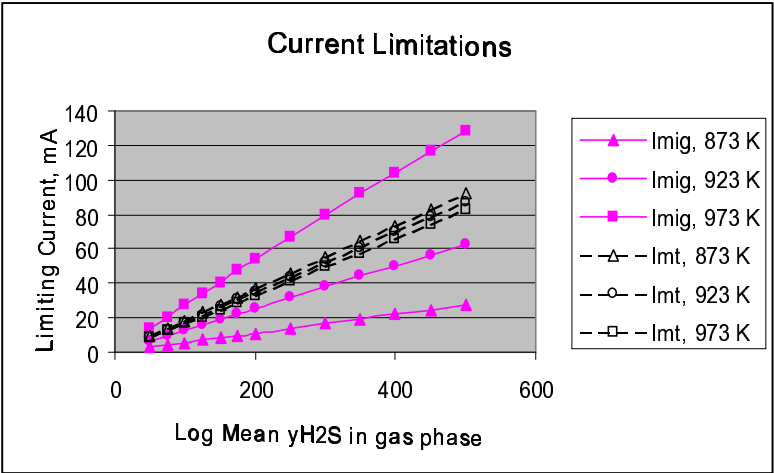


Figure 2. Mass Transfer Limited Current: Theoretical Limiting Migration Current vs. Bulk Mass Transfer Limited Current--both as functions of temperature and average H₂S concentration.

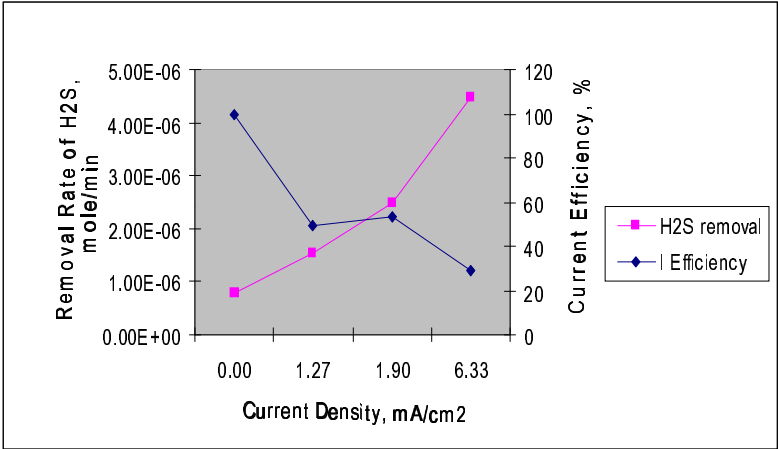


Figure 3. Removal Performance of Ca_(0.1)Y_(0.9)FeO₃ cathode run at 650° C.